Stress chains formation under shear of concentrated suspension

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Results comparing experiments on a model system of mono-disperse silica-particles with the numerical simulation of a highly concentrated suspension of spherical particles subject to a constant rate of strain are presented. Giant fluctuations of the shear stress and the first and second normal force difference are studied. Stress chain formation and evolution under shear are visualized in order to make the relation between the stress fluctuations and the suspension microstructure.

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INTRODUCTION

When submitted to a low stress, concentrated suspensions show an elastic response. By increasing the imposed stress, suspensions begin to flow with a plastic deformation. At higher shear stress they can shear thicken or even jam and stop flowing. Although these phenomena are of great importance for engineering applications such as extrusion or pumping, the basic understanding is very weak [1]. The simplest model to explain shear-thickening has been proposed by Boersma et al [2]: shear thickening appears when the hydrodynamic compressive forces are of the same order of magnitude as repulsive - either Brownian, steric or electrostatic - forces. The jamming transition can be continuous or discontinuous. In the latter case, when the suspension is forced to flow, we observe a fluctuation of the applied stress above a base line [3]. Two flow states are present: one plastic, which is the same as that in the plastic zone and the second flow state, where the stress is higher. The organization of the particles can be followed by experimental techniques such as confocal microscopy or neutron scattering [3-5]. While these techniques are used to follow the global organization of the particles under shear, they can not give information on the stress distribution in the suspension. Therefore, we turn to numerical simulation, using a dissipative particle dynamics (DPD) model [6], to study the organization of colloidal suspension under shear. The fluctuations of the tangential and normal stress are correlated to experimental data. The contacts between the particles are used to build a map of the stress occurring during the shear process. We can observe the formation of stress chains when highly concentrated suspensions are sheared.

JAMMING: EXPERIMENTAL VIEW

We use concentrated suspensions of silica particles of controlled roughness. These are prepared according to a Stöber-like synthesis [7] with an average size, measured by dynamic light scattering, of 1 μ m. Rheological measurements are made with a con/plate (Rheometrics* RFSII, 25 mm diameter). Confocal microscopy images are made with a Leica (TCS-4D, oil immersion 100 objective, equipped with an Ar-Kr laser), where the sample is sheared with a homemade cone-plate shear cell (radius 50 mm).

A concentrated suspension is sheared in a cone/plate geometry in order to measure the normal stress applied by the sheared suspension. We can then measure indirectly the anisotropy and spatial organization of the particles during the flow. This measurement is, however, difficult to perform: the jamming transition appears either at a high shear rate where the suspension is ejected from the gap or at a lower shear rate when the suspension is concentrated then a rapid drying occurs. We use rough silica particles, where the jamming transition occurs at a lower volume fraction and lower shear rate. The normal force fluctuates in phase with the tangential stress. The normal force fluctuations are negative at low volume fraction and low shear rate. At higher volume fraction, these are positive (see Figure 1).



FIGURE 1. Evolution of the tangential stress and normal stress as a function of the imposed shear rate and for different volume fractions (from left to right 42.5 %, 43 % and 44 % w/w). Data from reference 5.

Two images of the suspension, at rest, where the particles exhibit a liquid-like order, and after shear, above the jamming transition, where the particles are more heterogeneous, are given in Figure 2, as well as the Fourier transform of the centers of the particles. The volume fraction of the particles is 50 %. In the shear regime, where crystallites are observed, the suspension is highly shear thickening and becomes dilatant. However, as we are in the high concentration regime, we are not able to characterize the orientation of particle contacts.





FIGURE 2. Confocal images of suspensions at rest (A) and just after shear above the jamming transition (B). The Fourier transforms of the centers of the particles are on the left. Particles are in a mixture of water/glycerol at 23/77 *w/w*.

NEAR-JAMMING: SIMULATION RESULTS

The simulation approach used in this study is based on a hybrid dissipative particle dynamics approach (DPD) [6]. While interstitial fluid is modeled using DPD particles, the model explicitly incorporates lubrication forces when the particles are very close. For this paper, we consider a mono-size sphere suspension with solid volume fraction equal to 51 %. In the infinite Peclet Number regime, such systems have been shown to jam at small strains. The simulation approach used here can produce a jamming transition. However, as a consequence of modeling an interstitial fluid, with the compressibility of water, the usual jamming of a dense hard spheres suspension can be suppressed at lower shear rates. This effect may be controlled, allowing us to probe flow regimes very close to the jamming transition.

Similar fluctuations are observed with simulation as represented in Figure 3. Here giant fluctuations are found in the shear stress and first and secondary normal stress differences [1]. The first and secondary normal stress differences are generally positive, however, they occasionally showed negative fluctuations (not shown).



FIGURE 3. Shear stress (green), and first (red) and second (blue) normal stress differences. Results shown are normalized to the solvent fluid viscosity times shear rate.

Figure 4. shows evidence of the giant stress fluctuation in a highly correlated formation of stress chains



FIGURE 4. Stress chains visualization. Here, the suspension is straining to the right at the top and to the left on the bottom. The images are thresholded to only show the largest stresses with yellow indicate of the highest values. Note the lines are directed along the compression quadrant (oriented towards the bottom right). The image on the right is taken shortly after that on the left, indicating a minor change in microstructural organization at the onset of the stress jump.

In summary, we have shown, by computer simulation, evidence of transient stress chains that are oriented in the compression flow quadrant and rapidly reorganize themselves. These chains are responsible for the very large stress fluctuations. As found in experiment, these giant stress fluctuations appear near the jamming transition.

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REFERENCES

- 1. H. A. Barnes, Shear-thickening (dilatancy) in suspensions of non-aggregating solid particles dispersed in Newtonian fluids, Journal of Rheology 33, 1989 pp. 329-366.
- 2. W. H. Boersma, J. Laven and H. N. Stein, Shear thickening (dilatancy) in concentrated dispersions, AIChE 36, 1990, pp. 321-332.
- 3. D. Lootens, H. Van Damme and P. Hébraud, Giant stress fluctuations near the jamming transition, Phys. Rev. Lett. 90(17), 2003, 178301.
- 4. B.J. Maranzano and N.J. Wagner. Flow-small angle neutron scattering measurements of colloidal dispersion microstructure evolution through the shear thickening transition. Journal of Chemical Physics, 117, 22, 2002. pp. 10291-10302.
- D. Lootens, H. van Damme, Y. Hémar and P. Hébraud. Dilatant flow of concentrated suspensions of rough particles. Physical Review Letters, 95, 268302, 2005.
- 6. N. S. Martys, J. Rheol, 49(2), 401-424 (2005).
- 7. W. Stöber and A. Fink, J. Colloid Interface Sci. 26, 62-69.(1968).
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